5. Release Module

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Release is the rate that radioactive and chemical contaminants find their way into the environment. The Release Module handles liquid releases and releases from solid waste forms. Liquid releases are handled as a simple pass-through to the vadose zone or to the Columbia River. The solid forms are primarily from solid waste burial grounds including past-practice sites (pre-1988), active sites (post-1988), and the Environmental Restoration Disposal Facility. Other solid waste includes residual waste in the single- and double-shell tanks, naval reactor compartments, immobilized low-activity waste, the graphite cores of the retired production reactors, and concrete and cement waste associated with caissons and canyon buildings. The initial assessment included models for most of these releases to provide an estimate of contaminant release rate, as a function of time, to the vadose environment underlying the material disposal site. Release models for and naval reactor compartments are omitted from SAC because it is not anticipated they will release in the 1,000-year period of the initial assessment.

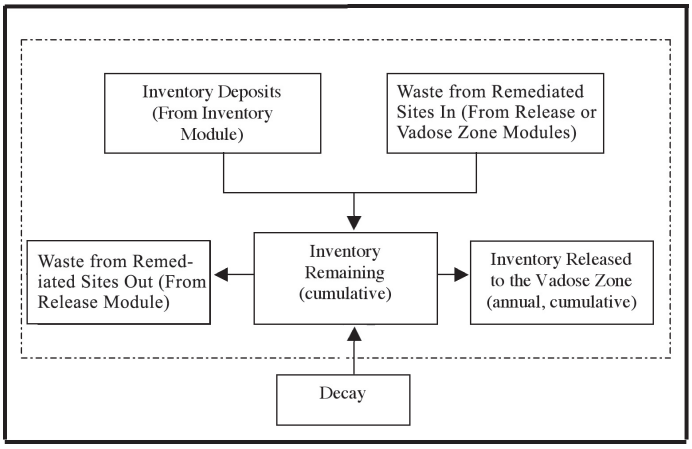

Figure 5.1. *Data inputs and outputs from the Release Module (VADER).*

The Release Module applies release models to waste inventory data from the Inventory Module and accounts for site remediation activities as a function of time. The resulting releases to the vadose zone, expressed as time-profiles of annual rates, become source terms for the Vadose Zone Module (Figure 5.1). Radioactive decay is accounted for in all inputs and outputs of the Release Module. The Release Module is implemented as the VADER (VADose zone Environmental Release) computer code.

Results

Releases to the environment at the Hanford Site began in 1944 and will continue until site closure. Table 5.1 shows a projection of the release of contaminants to the vadose zone from liquid discharge, solid waste

Table 5.1. *Deposited, released and residual inventory in waste deposits at site closure, 2050.*[(a),(b)]

Contaminant	Deposits (Discharged, Disposed, and Leaked)	Releases (to the Vadose Zone)	Remaining (in Waste Deposits)	% Remaining (in Waste Deposits)
Carbon Tetrachloride (kg)	814,000	4,250	810,000	99.5
Chromium (kg)	380,000	369,000	11,000	2.9
Uranium (Ci)	15,200	235	14,965	98.5
Iodine-129 (Ci)	78.3	7.38	70.9	90.5
Technetium-99 (Ci)	3,040	1,470	1,570	51.6
Plutonium-239 (Ci)	82,000	14,200	67,800	82.7
Cesium-137 (Ci)	1,060,000	280,000	780,000	73.6
Strontium-90 (Ci)	1,780,000	400,000	1,380,000	77.5
Tritium (Ci)	73,900	49,900	24,000	32.5

(a) Decay date for all radionuclides is 2050.

(b) Inventories exclude low-activity waste, naval reactor compartments, transuranic waste, high-level waste, spent fuel, and discharges to the Columbia River during reactor operations.

disposal, facilities, and unplanned releases for the entire site. Deposit, release and residual inventories are shown for the discharge and disposal sites. This information provides insight into the inventories that remain at waste sites in the relatively shallow surface disposal facilities. This summary table includes inventories discharged or disposed to the vadose zone prior to site closure; however, it does not include inventories that will not release to groundwater in the first 1000 years after site closure, e.g., immobilized low-activity waste, vitrification melters, and naval reactor compartments. Similarly, it does not include inventories scheduled for export, i.e., transuranic waste, high-level waste, spent fuel. Finally, it does not include discharges made to the Columbia River during the reactor operation era.

Quantities shown are based on annual means of the 25 realizations, decayed to the year assumed for site closure, 2050 AD, and summed over the entire operational era of the Hanford Site. Observations based on results from the Release Module include the following:

Waste forms for the Release Module include liquid releases, cement contaminated soil, saltcake residuals in tanks, and reactor blocks.

- The more highly sorbed contaminants including uranium, plutonium, cesium-137, and strontium-90 are held in waste deposits by their affinity for waste materials and sediments. The relatively high inventories retained in waste deposits are a result of their disposal in solid waste. Future disposals of uranium are assigned to stabilized waste forms and/or high integrity containers. Both create cementitious host environments that greatly retard release.

- Releases of the more highly sorbed contaminants including plutonium, cesium-137, and strontium-90 are a function of liquid discharges that occurred relatively early in Hanford Site history, (i.e., PFP waste streams containing plutonium sent to cribs and trenches, and tank wastes containing cesium-137 and strontium-90 sent to cribs and trenches during the mid-to-late 1950s). Hence, these inventories entered the vadose zone immediately upon discharge.

- Non-radioactive contaminants and radioactive contaminants with long half-lives are not influenced significantly by decay in Table 5.1; however, cesium-137, strontium-90 and tritium inventories are all influenced. Several million curies of each of these contaminants were discharged or disposed. Decay half-lives of 30.07, 28.78, and 12.32 years respectively for cesium-137, strontium-90 and tritium greatly reduce the inventories by the time of site closure. The remaining inventory of tritium at the time of site closure is highly related to the inventory assigned to the commercial low-level waste site, not to DOE wastes.

- Significant fractions of chromium, tritium, and technetium-99 are shown as released from waste sites, and, hence, as having entered the vadose zone. Each of these contaminants is highly mobile in the environment, and the large releases are indicative of liquid discharge inventories and unplanned releases (e.g., leaks) that were carried into the vadose zone immediately upon discharge. Inventories of tritium and technetium-99 shown as remaining in waste deposits are in solid waste disposal sites, often in cementitious waste forms.

- Iodine-129 can be mobile in Hanford vadose zone and aquifer sediments; however, relatively low releases have been simulated because most of these inventories are represented in the current inventory as

No glass waste forms were modeled in the initial assessment because negligible releases are expected from this waste form for the first 1,000 years after disposal.

residing in solid waste and not liquid discharges. Recent and future solid waste disposals of iodine-129 involve cementitious waste forms, (e.g., grout), or high integrity concrete containment, or both. Levels of iodine-129 contamination in liquid and atmospheric waste streams are under review, and the amount of iodine-129 retained in waste deposits will be revised downward.

- Chromium discharges and disposals occur on the river corridor and central plateau respectively. Much of the chromium associated with reactor operations is omitted from this discussion of release because it was discharged to the Columbia River a few hours after discharge from the single-pass reactors to retention basins. (Note, discharges to the river are included in the initial assessment.) However, some of the cooling water discharges were made to trenches, especially from the 100 K Area reactors, and therefore, a substantial quantity of chromium is reported as discharged to the vadose zone. The majority of the chromium inventory shown as retained is in the form of tank waste residuals at the time of site closure.

- Because of the need in the initial assessment to represent carbon tetrachloride release within the construct of an aqueous-phase (water) model, the release model selected was the solubility-limited desorption model. Carbon tetrachloride has a relatively low solubility in water, and the model tracks the mass of contaminant still available for release, so a large inventory is reported as remaining in the waste deposit. However, all of the carbon tetrachloride was released to the vadose zone. The 99.5% reported as 'remaining in the waste deposit' should be interpreted as 'released to the vadose zone but not the aquifer'. The simulated release, (i.e., 4250 kg), is lower than the field program's low extreme of estimated carbon tetrachloride release to the aquifer, (i.e., 5250 to 15,740 kg). The simulated release is also less than the field program's estimate of recovered carbon tetrachloride by the pump-and-treat remedial action, 5820 kg (Rohay 2002).

A great deal more information can be drawn from SAC simulations. Release data can be retrieved from SAC simulations to reveal the relative roles of release in each of the operation areas, between the central plateau and the river corridor, between tank wastes and all other Hanford Site waste types. Future efforts will make more use of the release results as analysts seek to understand the movement or propagation of inventory through the release model into the vadose zone, through the vadose zone into the groundwater, and through the groundwater into the river environment.

The summary information provided above for the entire Hanford Site is an overarching view of release. A great deal more information can be drawn from SAC simulations. Release data can be retrieved from SAC simulations to reveal the relative roles of release in each of the operation areas, between the central plateau and the river corridor, between tank wastes and all other Hanford Site waste types. Future efforts will make more use of the release results as analysts seek to understand the movement or propagation of inventory through the release model into the vadose zone, through the vadose zone into the groundwater, and through the groundwater into the river environment.

Conceptual Model

Key features that restrict the rate at which contaminants can be transported from the engineered system to the vadose zone are waste containment barriers, surface covers, and liners and leachate collection systems.

Waste containment facilities have a number of features that influence the rate at which contaminants can be released from waste. Those features are illustrated in Figure 5.2. The waste may be placed in a trench or reside in a tank. The trench, tank or other engineered structure may have features that serve as barriers preventing infiltrating water from making contact with and transporting contaminants from the waste to the vadose zone. Waste inside an engineered structure (e.g., trench) may also be contained in a waste package (e.g., a metal drum or high-integrity concrete container). The drum or concrete container acts as an additional barrier preventing transport of the contaminants from the waste. Major containment materials for Hanford waste are concrete, steel, and bituminous layers and coatings. The stability and permeability of containment materials change over time. Time affects which features dominate the water or contaminant migration in containment materials. Surface covers on an engineered system and liners (geomembrane and geosynthetic) and leachate collection systems at the bottom further restrict infiltrating water from transporting contaminants to the vadose zone. Surface covers are particularly important because migration of infiltrating pore water may be limited as long as the cover maintains its integrity.

Individual waste sites have one or more of the features shown in Figure 5.2. However, none of the waste sites in the initial assessment had all of the features in the conceptual model.

A number of key processes govern how much contaminant at any given time is released from the waste to the infiltrating water. One process is the affinity of contaminants to be retained by the waste (e.g., sorption to soil or waste material). Another process is the ability of waste to dissolve, and in some cases, to form new precipitates allowing some contaminants to be released to the infiltrating water while others remain trapped in the precipitated solids. Release from the waste may also be limited by the solubility of the contaminant in the infiltrating water.

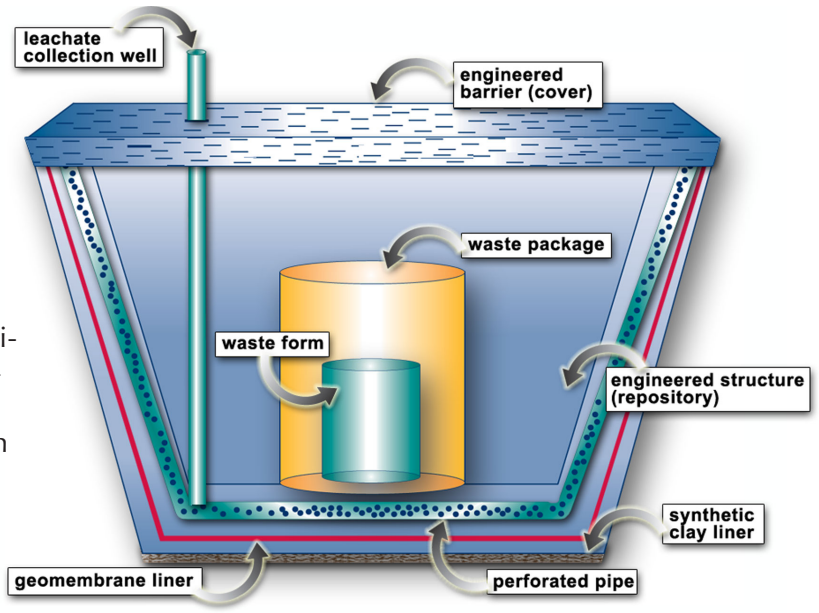

Figure 5.2. *Basic features of a waste containment facility.*

Water infiltrating an engineered system may contact and react with fill materials (e.g., soil, basalt, or grout), containment materials in various states of degradation, and different types of waste. Reaction with these materials will change the water chemistry, and physical and hydraulic properties over time. The water composition, pH, and redox state at any given time will influence the extent to which these processes influence contaminant release from the waste.

Implementation Model

The initial assessment took advantage of existing release models.

The initial assessment simulation begins in 1944 with an uncontaminated site. It accounts for annual inventory deposits and for remedial action transfers of waste to sites such as the Environmental Restoration Disposal Facility. The Release Module accounts for releases that occurred in the early years of Hanford Site operations and those that may be expected while the Hanford Site is remediated over the next several decades, in addition to long-term releases that continue until all inventory is released. The Release Module relies on several sources of input (Figure 5.3). Input from the Inventory Module includes contaminant mass (for chemicals) and activity (for radionuclides) deposits. Some of the release models (i.e., soil-debris, cement) require site or waste feature information (i.e., site cross-sectional area, site volume or waste surface area or volume). Recharge rate

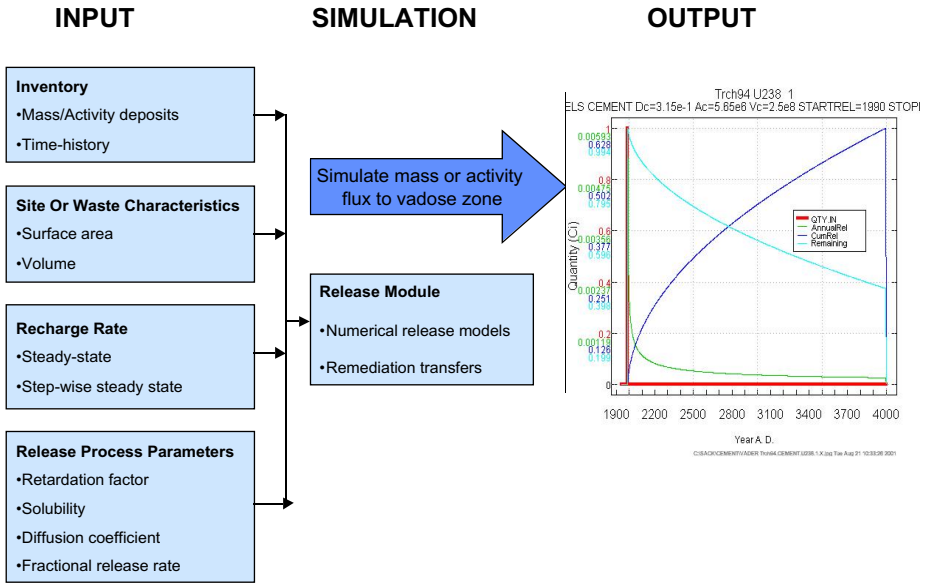

Figure 5.3. *Release implementation model for the initial assessment.*

is an important parameter to the salt cake and soil-debris models. Key process parameters are retardation factor (soil-debris model), solubility (soil-debris and salt cake models), diffusion coefficient (cement model) and fractional release rate (reactor block model).

A number of assumptions were made in the implementation model that result in a simplification of the conceptual model:

- Beyond the timing and magnitude of infiltration rates, the effects of waste containment due to packaging or engineered structures on contaminant releases from waste were not considered. Waste was allowed to release from the moment the inventory was placed in waste sites.

- Contaminant release from surplus production reactor waste cores was assumed to occur according to a simple linear fractional release rate, without taking account of the complex features of the core and their influence on contaminant release.

- Contaminants were released from tank waste assuming all the waste was salt cake and without accounting for the much lower release rates likely to occur for contaminants associated with tank waste sludge or hard heel.

- Waste sources requiring the application of models to simulate the release of contaminants from glass and naval reactor compartments were excluded because of their negligible influence on the overall output of the initial assessment, during the first 1,000 years after site closure (i.e., from 1944 to 3050).

- Carbon tetrachloride released from 200 West Area waste sites was modeled as being released to groundwater at the solubility limit of carbon tetrachloride in water. It was not modeled as a dense nonaqueous phase liquid in the vadose zone, and atmospheric losses and remedial actions were neglected.

- All release model parameters except infiltration were treated as stochastic over the suite of realizations and invariant within each realization over the full simulation period.

- Infiltration was varied over time to represent changes in land surface conditions (e.g., infiltration rates for pre-Hanford, operations, and surface barrier conditions, but infiltration time-profiles were the same for all realizations.

Processes that influence the release of contaminants from the waste to the percolating waters include adsorption, diffusion, solubility, and dissolution/ precipitation. How much these processes affect contaminant release from waste depends on the changes to the infiltrating water that contacts the waste over time.

The soil-debris model was applied to solid waste burial grounds and low-level waste burial grounds.

- Liquid and river releases were treated as instantaneous releases to the vadose zone and the Columbia River, respectively.

- Remediation transfers were treated as if completed within 1 year, although in practice they could require several years.

The Hanford Site also had releases of large volumes of contaminated water directly to the ground in trenches, ponds, and cribs. These releases are modeled as an immediate transfer to the Vadose Zone Module.

Uncertainty. In the SAC simulations, uncertainty is captured by expressing contaminant inventories and numerical model parameters in terms of statistical distributions. Each realization of the initial assessment used sample parameter values for randomly distributed variables such as: bulk soil density, soil moisture content, sorption coefficient, salt cake density, and cement diffusion coefficient. Other model parameters were held to constant values over all realizations.

Numerical Models

The waste in each of 533 waste sites, some of which are aggregate sources, was assigned to one or more of five numerical models within the Release Module (Table 5.2). The state-of-knowledge for each model used in the initial assessment is summarized in the following paragraphs.

Soil-Debris Model. The soil-debris model has two regimes: solubility-driven and desorption-driven. When desorption would yield a concentration greater than the solubility limit, it releases the maximum quantity the water can dissolve based on the contaminant solubility coefficient and recharge rate. When the mass or activity of an contaminant is less than the capacity of infiltrating waters to dissolve, the quantity released depends on the infiltration rate and soil characteristics.

In general, infiltration rates applied were those used in vadose zone modeling, which vary based on current or predicted site conditions (e.g., soil type, presence or absence of vegetation or a cover) (Fayer et al. 1999; Riley and Lo Presti 2001) and the precipitation record of the Hanford Site.

Values of aqueous solubility for contaminants were derived from experimental measurements or estimated based on geochemical calculations

Table 5.2. *Summary of waste sources included in the release numerical models.*

Numerical Model	Waste Source
Liquid	Single-shell tank past leaks and future losses,[a] unplanned releases,[b] trenches,[c] cribs,[c] drain/tile fields,[c] radioactive process sewers, French drains, retention basins, ponds, ditches, sumps, sand filters, injection/reverse wells, storage tanks, diversion boxes, catch tanks, valve pits, settling tanks, receiving vaults, and neutralization tanks.
Soil-Debris	Unplanned releases,[b] burial grounds, laboratories, storage, stacks, landfills, and decommissioned reactor compartments [d].
Cement	Process units or plants, control structures, grouted waste in storage tunnels, landfills, grouted waste in trenches, waste in HICs (high integrity concrete).[e]
Salt Cake	Single-shell tank[a] and double-shell tank[f] solid residuals.
Reactor Block[g]	Surplus production reactor graphite cores.

(a) Releases from single-shell tanks have been modeled using a combination of liquid and salt cake models. Releases include past tank leaks, liquid released during retrieval, and contaminant release from dissolution of residual solids after waste retrieval is completed.
(b) Modeled as initial liquid release, release from a surface-contaminated soil or a combination of both.
(c) Radionuclides from 216-Z-1A drain/tile field, 216-Z-9 trench, and 216-Z-18 crib are modeled as liquid release. Carbon tetrachloride from these sites has been modeled using the soil-debris model.
(d) Several chemicals and radionuclides (chromium and technetium-99) are found in reactor compartments. Following corrosion of the reactor hull, these contaminants would be released using the soil-debris model operating in the sorption- or solubility controlled mode.
(e) Waste contained in concrete vaults or other cement waste forms.
(f) Double-shell tanks are assumed not to leak before and during retrieval. Release of contaminants from residual solids (i.e., salt cake, sludge, and hard-heel) is assumed to be stoichiometric using the salt cake model. Stoichiometric are released based on the solubility of nitrate in tank supernatant.
(g) Release of B reactor core contaminants occurs entirely in the 100 Area. Following a specified period of time (75 years after issuance of SPR-EIS ROD, 2067), remaining inventories for all other reactor cores are moved to a 200 West Area burial ground (218-W-5), where release continues using the reactor block model.

(e.g., using the MINTEQA2 computer code [Allison et al. 1991]). Where the solubility of an contaminant was unknown, the aqueous solubility was fixed at an arbitrarily high default value forcing the soil-debris model to operate in the desorption-controlled mode. Data from Fayer et al. (1999), Peterson et al. (1996), and Schalla et al. (1988) were used to provide bulk density and volumetric moisture content values for use in soil-debris model simulations.

The salt cake release model was applied to solid waste remaining in high-level waste tanks following retrieval of the majority of waste for disposal.

Salt Cake Model. Contaminants are assumed to be contained within a waste matrix known as salt cake, which is comprised of salt cake, sludge, and hard heel without differentiation. The waste is assumed to be homogeneously distributed throughout the tank, and the salt cake matrix is assumed to be composed mostly of nitrate salts. This salt cake is assumed to contain the contaminants that dissolve in percolating waters congruently with the matrix. The salt cake model consists of a simple analytical solution containing a term for infiltration, matrix (nitrate) solubility, and the cross-sectional area of the waste source (i.e., single-shell or double-shell footprint. Recharge rates for the salt cake model are the same as described for the soil-debris model. Sources of data on recharge and cross-sectional area for this model can be found in Riley and Lo Presti (2001). Because the Inventory Module provides the salt cake matrix of each tank as a volume, a salt cake matrix density value is required to convert tank waste volumes to equivalent masses. Applications of the salt cake model for the initial assessment used a statistically derived value of density for tank solids (Chen et al. 1998) based on tank characterization data. Typical density values are around 1.5 grams per cubic centimeter.

Cement Model. The cement model is applied to waste that has the characteristics of cement. The total external surface area and the volume of the waste must be known. The ratio of area to volume is assumed to be constant. The most important term in the model is the diffusion coefficient. It describes the rate at which the contaminant migrates from the interior of the cement form to its surface. Diffusion coefficients for selected radionuclides have been determined for unsaturated conditions (Mattigod et al. 2001). Diffusion coefficients not provided by Mattigod et al. (2001) were obtained from Serne et al. (1992) and were based on saturated moisture conditions.

The reactor block model is used to simulate the release of contaminants from decommissioned production reactors on the Hanford Site.

Reactor Block Model. The reactor block model is used to simulate the release of contaminants from decommissioned production reactors on the Hanford Site. The analytical solution is simple, consisting of only a mass and fractional release term. These release rates have been calculated from experimental leach rates (White et al. 1984, U.S. Department of Energy 1989) and Hanford reactor configurations.

History Matching

The ability to "history match" simulated release of contaminants from Hanford waste sources to the vadose zone has been impossible because no field data are available. Because direct history matching of the release

models is not possible, history matching of other modules, such as the Vadose Zone Module, must be used to determine the adequacy of the release models. Future efforts will evaluate the performance of the combination of inventory, release, and vadose zone modules (i.e., release to aquifer water table) against aquifer contaminant plume data sets. These future history matching efforts will focus on tritium, strontium-90, technetium-99, iodine-129, and uranium contaminants.

The Release Module provided acceptable results for this initial assessment. The numerical model can be improved as additional data about waste inventories and types are incorporated into the model.